Comment on "Competition between helimagnetism and commensurate quantum spin correlations in $LiCu_2O_2$"

In a neutron scattering investigation of $LiCu_2O_2$ Masuda *et al.*[1] reported the direct observation of an incommensurate (IC) magnetic structure below 22 K. Though this study confirms similar indirect IC observations[2, 3, 4] pointing to the presence of frustrated magnetic interactions they deserve now more detailed work to elucidate the microscopic origin of that frustration. We will show that the adopted antiferromagnetic (afm) double-chain (DC) Heisenberg model [1, 2, 3] (Fig. 1a) suggests an unrealistic frustation scenario for $LiCu_2O_2$. It should be replaced by a ferromagnetic(fm)-afm frustrated *single*-chain model (Fig. 1b).[4, 5] Based on electronic structure (LDA) and cluster calculations as well as a phenomenological analysis of magnetic data, we arrive at opposite estimates compared with Masuda *et al.* [1] with respect to the magnitude/sign of the main couplings. The controversy concerns the following main points:

(i) Most importantly, the signs of the n.n. inchain exchange J_1 are opposite: afm +1.68 meV in Ref. 1 vs. fm -11± 3meV in our analysis.[6, 7] For CuO_2 chains with Cu-O-Cu bond angles γ near 94° as in Li_2CuO_2 (with fm inchain order), according to the Kanamori-Goodenough rule and to the fm direct Cu $3d$-O $2p$ exchange, a total *fm* J_1 <0 can be expected. However, its magnitude is sensitive to the competition with a γ-dependent afm contribution to J_1.[4] Hence, to simplified distance-only based suggestions [1] that $|J_1| \gg J_2$, do note hold here.

(ii) We found the nnn inchain coupling J_2 afm (generic for CuO_2 chains), i.e. frustrated with fm J_1 and any J_{DC}. Moreover we estimated $J_2 \sim |\ J_1\ |$. However, the important source of frustration J_2 is ignored in Ref. 1.

(iii) A dominant interchain coupling $J_{DC} \approx 5.8$ meV is claimed by Masuda *et al.* whereas from our LDA analysis a tiny $J_{DC} \sim 0.5$ meV only follows. It can be neglected to first approximation. The weak J_{DC} is caused by the tiny interchain (DC) overlap of the predominant O $2p_{x,y}$ orbitals of the CuO_4 plaquettes forming the CuO_2 chains. Note, that if J_1<0, the DC is *unfrustrated* for J_2=0.

With $J_1 \sim$-11meV, we explain also the measured magnetic susceptibility $\chi(T)$ (Fig. 1) and the afm Curie-Weiss constants Θ_{CW}. [2] Approximating the main couplings between two nn-chains $J_\perp$ in the ab-plane in mean-field theory and the inchain couplings exactly in large clusters, we derive for the bulk value $\Theta_{CW} \approx \Theta_{CW,1D} - zJ_\perp/4k_B$, where z=2 is the number of interchain n.n.'s. From cluster studies we obtained an afm $\Theta_{CW,1D} \approx$ -42K for single chains with frustrating afm J_2 >0 and $\alpha \equiv J_2/J_1$=-1.1. With $J_\perp \approx$5.7meV [4] one arrives at Θ_{CW}= -75K close to experimental values ≈ -80 to -90K.[2, 8]

Finally, we note that Masuda et al. [1] argue that their propagation vector ζ would contradict our J ratio: α=-1/(4cos(2$\pi\zeta$)). However, this simple expression is valid for single-chains with classical spins $s \gg 1$. In our case with s=1/2 quantum fluctuations[9], interchain coupling[10], and spin-anisotropy do affect α strongly.

To conclude, the application of the afm DC-model of Ref. 1 to $LiCu_2O_2$ is not justified whereas the proposed frustrated single-chain model with fm J_1 and afm J_2 couplings is consistent with the experimental data and the generally accepted CuO_2-chain physics.

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[1] T. Masuda *et al.*, Phys. Rev. Lett. **92**, 177201 (2004).
[2] S. Zvyagin *et al.*, Phys. Rev. B **66**, 064424 (2000).
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[5] The observed propagation vector $q = (0.5, \zeta = 0.174, 0)$ measured in standard units $2\pi/b$ of the crystal structure yields a pitch angle $\Phi = \pm 2\pi\zeta + 2\pi n$, n=0,1,2,..., within the "fm" interval $-\pi/2 \leq \Phi \leq \pi/2$ [9]. However, within the system of coordinates adopted in Ref. 1 with a lattice constant of $b/2$ there are two solutions $\Phi_{DC} = \pm\pi(n \pm \zeta)$ with "afm" pitch angles for odd (even) n, respectively, depending on the sign of J_{DC}.
[6] The error bars stems from uncertainties in the screening of bare fm interactions.[4]
[7] From mapping low-lying magnetic excitations of the 5-band Cu $3d$ O $2p$ extended Hubbard model (fitted to spectroscopic data) onto excitations of the Heisenberg model.
[8] Notice the "+"-sign convention of Θ_{CW} used in Ref. 2.
[9] R. Bursill *et al.*, J. Phys. C, **7**, 8605 (1995), Fig. 2.
[10] Strong interchain exchange may even prevent a helix for-

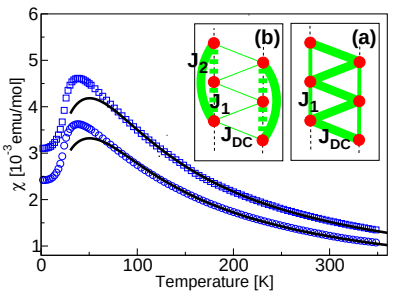

FIG. 1: Susceptibility of Heisenberg rings with $-J_1 = J_2$=8.2 meV, J_{DC}=0. N=16 sites, and Lande-Factors g_L= 2.24 and 2.0, respectively (full lines) compared with experiment (Ref. 1; □ magnetic field **H** ∥ c; ○ **H** ∥ (a,b). In the inset the DC scenario ((a),[1]) is compared with the single-chain one (b). Thickness of lines symbolizes the coupling strength. The empirical J-values are in accord with LDA and microscopic estimates [7]. Naturally, the finite cluster approach cannot describe the low-T behavior of $\chi(T)$.

mation, e.g. as in Li_2CuO_2 with fm inchain order below 8K.